

Decay of Nb^{94} and $\text{Nb}^{94m\ddagger}$

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The beta decay of 2.0×10^4 -yr Nb^{94} has been investigated by means of a scintillation spectrometer. The beta spectrum has an end-point energy of 470 ± 6 keV and yields a linear Fermi-Kurie plot when a shape factor of $p^2 + \lambda q^2$ ($\lambda = 1.75 \pm 0.15$) is applied. The shape factor indicates the transition is second-forbidden non-unique, in accord with its $\log ft$ value of 12. Additional measurements were made of electromagnetic radiations from both Nb^{94} decay and that of 6.6-min Nb^{94m} . No new transitions were observed, but more precise energy determinations of the gamma rays emitted in the decay of Nb^{94} were obtained. The results of this study further support previously proposed spin and parity assignments.

I. INTRODUCTION

PREVIOUS investigations¹⁻³ have shown that the ground state of Nb^{94} decays by beta emission with a half-life of 2.0×10^4 yr to the 1.57-MeV excited state of Mo^{94} . This state in turn is de-excited by the emission of cascade gamma rays of 0.70 and 0.87 MeV. Coulomb excitation experiments^{4,5} have demonstrated that the 0.87-MeV level has a spin and parity of $2+$, while the results of a recent study by Reich, Schuman, and Heath³ have shown the most likely spin and parity of the 1.57-MeV state of Mo^{94} to be $4+$. Figure 1 gives the basic decay scheme proposed by these authors for both the decay of Nb^{94} and the 6.6-min isomeric state, Nb^{94m} , lying 41 keV above the ground state. The results of further investigations reported here, which include more precise values for the various beta- and gamma-ray energies, are also incorporated in the decay scheme.

Reich *et al.*³ point out that it is somewhat unexpected to find the decay of the 6.6-min Nb^{94m} proceeding by an $E3$ transition to the ground state and they suggest the possibility of a low-lying intermediate level. Since the spins and parities of the two states of Nb^{94} as well as the 1.57-MeV level of Mo^{94} have been assigned on the basis of indirect evidence, it was felt that a further study was worthwhile, especially if it included a direct measurement of the spectrum shape for the beta decay of Nb^{94} . The maximum energy of this beta transition is approximately 0.5 MeV¹ which, together with its 2.0×10^4 -yr half-life, gives a $\log ft \approx 12$. This indicates that the beta transition can be characterized as either $\Delta I = 2(\text{no})$ or $\Delta I = 3(\text{no})$. In general, the two possibilities will produce differently shaped beta spectra. No previous spectrum-shape measurement has been reported.

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¹ D. L. Douglas, A. C. Mewherter, and R. P. Schuman, *Phys. Rev.* **92**, 369 (1953).

² R. P. Schuman and P. Goris, *J. Inorg. Nucl. Chem.* **12**, 1 (1959).

³ C. W. Reich, R. P. Schuman, and R. L. Heath, *Phys. Rev.* **129**, 829 (1963).

⁴ G. M. Temmer and N. P. Heydenburg, *Phys. Rev.* **104**, 967 (1956).

⁵ P. H. Stelson and F. K. McGowan, *Phys. Rev.* **110**, 489 (1958).

In this investigation we undertook a measurement of the spectrum shape in the beta decay of Nb^{94} and a survey of the low-energy electromagnetic radiations emitted in the decay of 6.6-min Nb^{94m} . In addition, more precise energy determinations of the gamma rays emitted in the decay of Nb^{94} were obtained.

II. INSTRUMENTATION AND SOURCES

A. Nb^{94}

The beta spectrometer consisted of two treated cylindrical Pilot-B plastic scintillators ($1\frac{1}{8}$ in. in diam $\times \frac{1}{8}$ in. thick) which were optically coupled with a thin layer (approximately 0.2 mg/cm²) of liquid containing the radioactive source.⁶ This scintillator sandwich was cemented with epoxy to a $1\frac{1}{2}$ -in.-diam $\times \frac{1}{2}$ -in.-thick Lucite light pipe. A coating of reflective paint was used on both the sandwich and light pipe to obtain good reflection and handling characteristics. The light pipe was mounted on an RCA No. 8053 photomultiplier (selected for low noise and high resolution) and the pulse-height spectrum was recorded with a multi-channel analyzer. A 3-in. \times 3-in. integral-mounted NaI(Tl) crystal placed in front of the beta detector served as a coincidence detector.

The spectrometer was calibrated by using internal-conversion electrons of In^{114m} (163 keV), In^{113m} (363 keV), and Ba^{137m} (624 keV) in coincidence with their

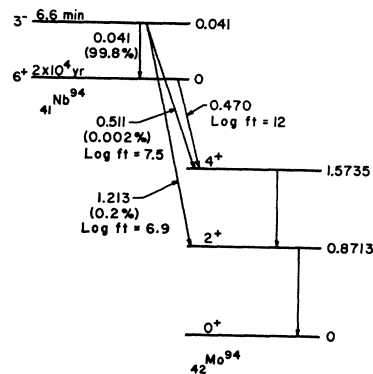


FIG. 1. Decay scheme of Nb^{94} and Nb^{94m} . All energies are in MeV.

⁶ R. E. Snyder and G. B. Beard, *Nucl. Instr. Methods* **26**, 31 (1964).

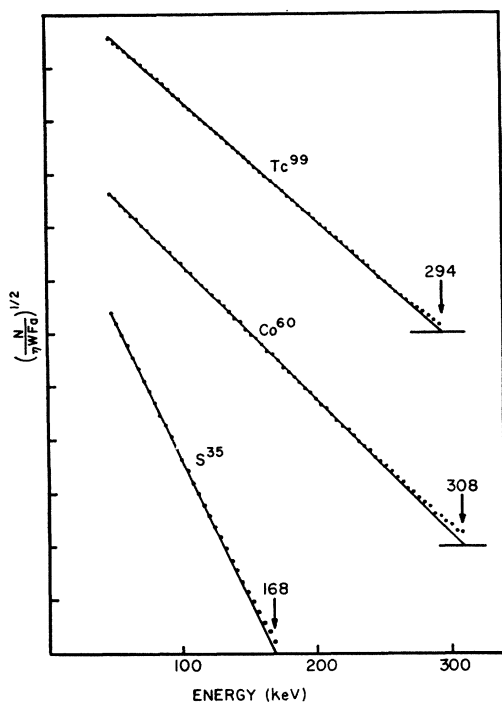


FIG. 2. Fermi-Kurie plots for S^{35} , Co^{60} , and Tc^{99} . A shape factor of $p^2 + 2.05q^2$ was used to make the Tc^{99} plot linear.

K x rays. The linearity was found to be better than 1%; previous work⁶ has shown that such a system is linear down to below 50 keV. The resolution (full width at half-maximum) of the Ba^{137m} K internal-conversion line was 10.5%.

The source of the 2×10^4 -yr Nb^{94} was produced by neutron irradiation of spectroscopically pure niobium metal powder.⁷ Approximately 1 mg of the Nb^{94} , in the form of niobium metal, was dissolved in a solution of HF and HNO_3 , which, with the addition⁶ of H_2O , glycerin, and $SnCl_2$, brought the total weight to approximately 20 mg. Only 1.1 mg of this solution was used to form the scintillator sandwich described earlier and the counting rate obtained from the Nb^{94} beta activity was low enough to avoid possible difficulties with accidental coincidences from either the Nb^{94} activity or with x rays from 12-yr Nb^{93m} present as a contaminant.

The gamma-ray spectrum was observed by means of a 12-cc Li-drifted germanium detector obtained from Nuclear Diodes Corporation, and the data were recorded with a multichannel analyzer. Comparison sources of Co^{60} , Cs^{137} , Mn^{54} , Y^{88} , Na^{22} , and ThC'' were used for energy calibration.

B. Nb^{94m}

To produce the 6.6-min Nb^{94m} activity, 5-mg samples of "Specpure" Nb metal were irradiated for periods of several minutes in a neutron flux of 4.5×10^{12} n/cm²

⁷ For details of the source preparation and purification, see Ref. 3.

sec at the University of Michigan Ford Reactor. The singles spectrum was taken using a xenon-filled proportional counter, and the data were recorded with a multichannel analyzer. The photon spectrum in coincidence with the 17-keV x rays of Nb was also obtained. In this case the x rays were detected by a second xenon-filled proportional counter whose output, after amplification, was fed into a single-channel analyzer. Pulses from the single-channel analyzer triggered a univibrator which in turn was used to gate the multichannel analyzer. The resolving time of the coincidence circuitry was about 3 μ sec.

III. EXPERIMENTAL RESULTS AND ANALYSIS

A. Nb^{94}

Before analysis, all beta spectra were corrected for the finite resolution of the spectrometer.⁸ Fermi-Kurie plots were constructed utilizing values from published tables⁹ with outer screening corrections¹⁰⁻¹² being applied in all cases.

To check the performance of the spectrometer, spectra of 5.2-yr Co^{60} ($\Delta I=1$, no), 87-day S^{35} ($\Delta I=0$, no), and 2.1×10^5 -yr Tc^{99} ($\Delta I=2$, no) were obtained. The latter two nuclei decay by pure beta emission and therefore coincidence measurements were not required for them. After corrections were made for background (less than 1%) S^{35} yielded a Fermi-Kurie plot linear down to approximately 50 keV with an end-point energy of 169 ± 2 keV. By using a shape factor¹³ of $p^2 + \lambda q^2$ ($\lambda = 2.05 \pm 0.15$) the Fermi-Kurie plot for Tc^{99} was also made linear above 50 keV and had an end-point energy of 294 ± 4 keV (p is the electron momentum, q the neutrino momentum, and λ a variable parameter). These results are all in good agreement with currently accepted values for these nuclei.¹⁴⁻¹⁶

The decay scheme of Co^{60} is similar to that of Nb^{94} , since Co^{60} decays almost wholly by beta emission to the 2.50-MeV state of Ni^{60} which then de-excites by the emission of cascade gamma rays of 1.17 and 1.33 MeV. By measuring the beta spectrum in coincidence with the gamma sum peak at 2.50 MeV the effect of Compton scattering of the gamma rays in the scintillator sandwich was avoided and the resultant Fermi-Kurie plot

⁸ D. E. Wortman and J. G. Cramer, Jr., Nucl. Instr. Methods **26**, 257 (1964).

⁹ Tables for the Analysis of Beta Spectra, (U. S. Government Printing Office, Washington, D. C., 1952), Natl. Bur. Std. Appl. Math. Ser. No. 13.

¹⁰ M. E. Rose, Phys. Rev. **49**, 727 (1936).

¹¹ C. Longmire and H. Brown, Phys. Rev. **75**, 264 (1949).

¹² C. Longmire and H. Brown, Phys. Rev. **75**, 1102 (1949).

¹³ E. J. Konopinski and M. E. Rose, in *Alpha-Beta- and Gamma-Ray Spectroscopy*, edited by Kai Siegbahn (North-Holland Publishing Company, Amsterdam, 1965), Vol. 2, p. 1357.

¹⁴ R. D. Connor and I. L. Fairweather, Proc. Phys. Soc. (London) **A70**, 769 (1957).

¹⁵ C. S. Wu, in *Beta- and Gamma-Ray Spectroscopy*, edited by Kai Siegbahn (Interscience Publishers Inc., New York, 1955), p. 345.

¹⁶ F. Wagner, Jr., and M. S. Freedman, Phys. Rev. **86**, 631 (A) (1952).

for Co⁶⁰ was found to be linear down to about 50 keV with an end-point energy of 308 ± 3 keV, a result in agreement with other recent work.¹⁷ Linear Fermi-Kurie plots for the three isotopes are shown in Fig. 2.

In the investigation of Nb⁹⁴, the beta spectrum was measured in coincidence with the gamma-ray sum peak at 1.57 MeV. With this arrangement the counting rate was about 10 counts/sec. Fermi-Kurie plots were made assuming an allowed transition, and first-, second-, or third-forbidden unique transitions with the appropriate shape factors,¹⁸ as well as a second-forbidden non-unique transition having a shape factor of $p^2 + \lambda q^2$.

The only linear Fermi-Kurie plot (see Fig. 3) was found for the shape factor corresponding to the second-forbidden nonunique transition ($\Delta I=2$, no) with a value of $\lambda=1.75 \pm 0.15$ and an end-point energy of 470 ± 6 keV. In particular, the shape factor corresponding to a second-forbidden unique transition ($\Delta I=3$, no) yields a distinctly nonlinear Fermi-Kurie plot, as shown in Fig. 3. Thus the beta spectrum of 2.0×10^4 -yr Nb⁹⁴ can be characterized by $\Delta I=2$, no, with an end-point energy of 470 ± 6 keV and a $\log ft$ of 12.

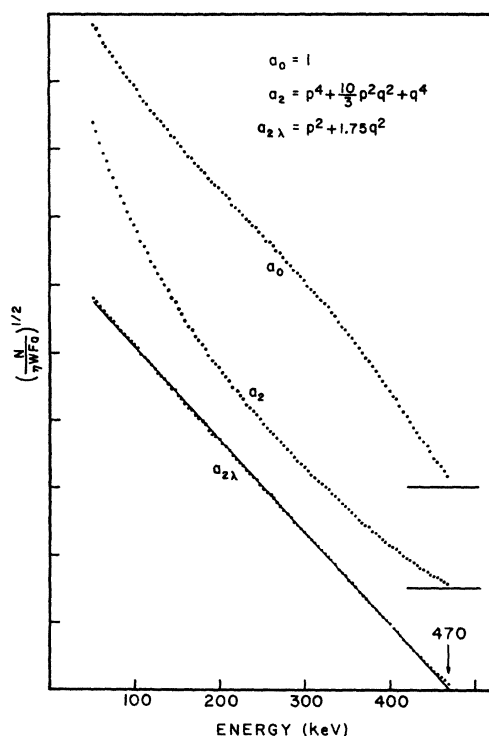


FIG. 3. Fermi-Kurie plots for Nb⁹⁴ assuming allowed, second-forbidden unique and second-forbidden non-unique shape factors. The statistical errors are about the size of the dots, except at the high-energy end of the spectrum.

¹⁷ F. Bonhoeffer, Z. Physik **154**, 62 (1959).

¹⁸ C. S. Wu, in *Alpha-Beta-, and Gamma-Ray Spectroscopy*, edited by Kai Siegbahn (North-Holland Publishing Company, Amsterdam, 1965), Vol. 2, p. 1373.

An analysis of the Nb⁹⁴ gamma-ray spectrum obtained using the solid-state detector did not show the presence of any new gamma rays, but did yield more precise values of 702.6 ± 0.5 and 871.3 ± 0.5 keV for the cascade gamma rays of Mo⁹⁴. The value of 871.3 keV is in good agreement with the recent result of Robinson *et al.*¹⁹ who find an energy of 871.1 ± 0.7 keV for the gamma ray emitted by Mo⁹⁴ following Coulomb excitation.

The spectrum also showed a sum peak having an energy of 1572.9 ± 1.0 keV (Reich *et al.*³ have placed an upper limit of 0.03% for the intensity of the crossover transition in agreement with earlier work²). A weighted average of the sum of the energies of the individual gamma rays and the value obtained from the sum peak yields an energy of 1573.5 ± 0.6 keV for the second excited state of Mo⁹⁴.

B. Nb^{94m}

The singles spectrum of the 6.6-min Nb^{94m} activity taken with the xenon-filled proportional counter showed only the peak due to the 17-keV characteristic K x rays of Nb. No evidence for other photons in coincidence with these x rays was seen when the output of the second proportional counter was used to gate the multichannel analyzer. It was estimated that any coincident photons lying in the range from about 4 to 40 keV would have been detected with fair efficiency.

IV. DISCUSSION

The results of this investigation further confirm the spin and parity assignments proposed by Reich *et al.*³ in the decay of the two Nb⁹⁴ activities. In particular, the beta decay of Nb⁹⁴ has been shown to proceed via a second-forbidden non-unique transition ($\Delta I=2$, no) confirming the spin and parity assignment of $6+$ to the ground state of Nb⁹⁴, based on the assumption that the 1.57-MeV state of Mo⁹⁴ is $4+$. In addition, the result of the survey of the low-energy photon spectrum in the decay of Nb^{94m} indicates that there is no intermediate level between the 41-keV isomeric state and ground state of Nb⁹⁴, thereby supporting the 3- assignment to the 41-keV state of Nb⁹⁴. However, it should be pointed out that the problems involved in making configuration assignments to the two states in Nb⁹⁴ still exist.³

Since the beta-spectrum shape in the decay of Nb⁹⁴ was observed, the end point could be determined with fair precision which, together with the improved gamma-ray energy measurements, allows the various energy differences in the decay scheme to be specified more accurately. An end-point energy of 470 keV for the beta decay from the ground state of Nb⁹⁴ implies end-point energies of 0.511 and 1.213 MeV for the beta decay of the 6.6-min Nb^{94m} to the 1.57- and 0.87-MeV

¹⁹ R. L. Robinson, P. H. Stelson, F. K. McGowan, J. L. C. Ford, Jr., and W. T. Milner, Nucl. Phys. **74**, 281 (1965).

levels of Mo⁹⁴, respectively. With the beta branching ratios found by Reich *et al.*³ the $\log ft$ value for the beta transition to the 1.57-MeV level is 7.5, and 6.9 for the transition to the 0.87-MeV state.

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Differential Scattering of Neutrons from ²⁰⁸Pb at Resonant Energies*

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Measurements are reported on differential elastic scattering of neutrons of a few keV energy spread from 99.75% ²⁰⁸Pb in the energy region 0.715 to 1.761 MeV. The results include absolute differential cross sections at 20 resonant and 9 nonresonant energies. A least-squares phase-shift analysis provides the resonance parameters and reduced widths of 21 resonances. The observed $s_{1/2}$ potential-scattering phase shifts are in agreement with those expected from a type of phenomenological potential which reproduces the single-particle bound states of ²⁰⁹Pb. Parity assignments and widths of the $J = \frac{1}{2}$ resonances are in disagreement with the prediction for 5 intermediate states arising from two-particle one-hole neutron excitations. There are, however, a number of resonances with widths corresponding to several percent of the single-particle limits which are likely candidates for some sort of intermediate states.

INTRODUCTION

THE interaction of a nucleon with a closed-shell nucleus is expected to give a relatively simple spectrum. States near the ground states should be essentially single-particle in character; the nucleon is loosely bound to the relatively stable core. At somewhat higher energies short-lived intermediate states should appear for which the core is in simple states of excitation. For ²⁰⁸Pb plus a neutron the low-lying states seen in (*d,p*) stripping¹ can be described in terms of the shell model.² Neutron scattering from ²⁰⁸Pb shows up a number of resonances with widths consistent with those expected for intermediate states.³ Of the over 100 resonances in the neutron total cross section up to 2.0-MeV neutron energy, about one-third have widths greater than 3 keV and are sufficiently isolated so that their J value can be assigned.⁴⁻⁷ Shakin⁸ has compared the widths of 6 observed $J = \frac{1}{2}$ resonances⁶ with widths

calculated under the assumption that these were $s_{1/2}$ intermediate states arising from two-particle one-hole neutron excitations in ²⁰⁹Pb. He found order of magnitude agreement. Of course, from total cross-section data, one in general deduces only J values and total widths. Angular distributions are necessary to assign l values and reduced neutron widths. This paper is concerned with the experiment designed to obtain such angular distributions, with the partial-wave phase-shift analysis of the differential scattering, and with the conclusions to be drawn from such analysis.

EXPERIMENT AND RESULTS

In order to measure differential cross sections at the resonances in ²⁰⁸Pb special apparatus had to be designed.⁹ In the first place, the required ~ 3 -keV energy spread is about a factor of 5 to 10 times smaller than was customary in this type of work. Furthermore, the quantity of ²⁰⁸Pb separated isotope available ($\sim \frac{1}{2}$ mole) was at least a factor of 2 less than that used in such previous experiments with light elements at resonant energies.⁹

Figure 1 is a drawing of the apparatus built to do this job. The neutron source is the ⁷Li(*p,n*)⁷Be reaction pro-

* Research sponsored by the U. S. Atomic Energy Commission under contract with the Union Carbide Corporation.

¹ P. Mukherjee and B. L. Cohen, *Phys. Rev.* **127**, 1284 (1962).

² J. Blomqvist and S. Wahlborn, *Arkiv. Fysik* **16**, 545 (1960).

³ C. Shakin, *Ann. Phys. (N. Y.)* **22**, 373 (1963).

⁴ R. E. Peterson, R. K. Adair, and H. H. Barschall, *Phys. Rev.* **79**, 935 (1950).

⁵ R. M. Wilenzick, G. E. Mitchell, K. K. Seth, and H. W. Lewis, *Phys. Rev.* **121**, 1150 (1961).

⁶ J. L. Fowler and E. C. Campbell, *Phys. Rev.* **127**, 2192 (1962).

⁷ J. A. Farrell, G. C. Kyker, Jr., E. G. Bilpuch, and H. W. Newson, *Phys. Letters* **17**, 286 (1965).

⁸ J. L. Fowler, *Bull. Am. Phys. Soc.* **7**, 576 (1962).

⁹ M. D. Goldberg, V. M. May, and J. R. Stehn, Brookhaven National Laboratory Report No. BNL 400, 1962, 2nd ed. (unpublished).